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RADIONUCLIDE FRACTIONATION IN AIR-BURST DEBRIS

by

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ABSTRACT

Radiochemical data from fractionated samples of air-burst debris are correlated logarithmically. The correlation slopes are measures of volatility. These are compared with slopes observed in the cases of high-yield surface bursts and with thermodynamic calculations of volatility.

SUMMARY

Radiochemical data from fractionated samples from 15 airbursts provided a source of equivalent-fission values for 24 radionuclides. The airbursts ranged in yield by a factor of over 300. The equivalent-fission values were converted to ratios based on Zr^{95} and these ratios were correlated logarithmically. The correlation slopes were found to be relatively insensitive to yield and their values permitted the placement of the radionuclides on a scale of volatility. There were found several groups of radionuclides which did not fractionate from each other. The nuclides Cs^{137} and Ba^{140} had essentially the same slopes for air bursts as for high-yield surface bursts, but Mo^{99} and Np^{239} behaved more volatilely in the air bursts. Finally, the volatility inferred from fractionation behavior correlated well with that based on thermodynamic calculations.

INTRODUCTION

When one considers world-wide fallout, air bursts are of much greater significance than surface bursts for two reasons. First, because a much greater quantity of fission products has been produced by air bursts than by surface detonations and secondly, because the entire amount of debris from an air burst goes into world-wide fallout, as compared to some small but unknown fraction for a surface burst.

Although a large amount of information has been collected on air-burst debris, relatively little has appeared in the unclassified literature. A few remarks on the nature of air-burst debris are therefore in order by way of background. The particles from air bursts are almost exclusively spherical, although occasionally particles are found which consist of two spheres stuck together. The particles are small, rarely occurring larger than 20 μ in diameter. Lapple has estimated that these particles can grow only to 0.3 μ by condensation and are probably no smaller than 0.03 (μ).[1] Therefore either the larger particles must have been formed by the coalescence of small particles when the debris was still in the molten state or they were formed from debris which was never completely vaporized. In either case they probably consist predominantly of particles formed at the earlier times.

The size-frequency distribution has been measured down to 0.05 micron diameter and found to be approximately lognormal with a modal diameter less than one micron.* The densities of the particles vary from 3 to 4.3 g/cc.[2] Their colors may be colorless, gold, yellow, orange, red, brown, green or black. Their chemical composition is usually a mixture of the oxides of iron, aluminum, uranium and plutonium. Radiochemically, their specific activity is much greater than that found in surface-burst debris, as would be expected. Thus, while the more active particles from surface bursts contain of the order of 10^{14} Zr^{95} equivalent fissions per gram,** air burst particles may have specific activities from 10^{4} to 10^{7} times higher. Benson and coworkers have

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* Leventhal, L., Tracerlab, Inc., Private communication.

**An equivalent fission of a fission product or induced activity in any given event is the ratio of the total quantity of product to the total number of fissions and thus represents the average quantity of product formed by one fission.

recently determined that in larger particles from air-burst debris Zr^{95} content varies as the 3.1 to 3.4 power of the diameter, while Ba^{140}-La^{140} content varies as the 2.3 to 2.6 power.*

Most of the samples studied have shown a relatively small degree of fractionation. However, since most samples were collected with precautions to obtain as representative a collection as possible, one should not leap to the conclusion that air-burst debris is fractionated to only a small extent. A large number of samples have also been obtained which showed large departures from the representative composition. The analytical data from these samples provides a wealth of material for analyzing fractionation in air-burst debris.

It is helpful to draw a distinction between potential and actual fractionation when considering the extent to which air-burst debris is fractionated. By potentially fractionated debris we mean debris for which the radiochemical composition varies among particles, whether because of their size, density, macroscopic composition or history in the fireball, even though these particles are intimately mixed so as to provide a representative composition for any given cubic foot of cloud. For example, the work of Benson, et al. cited above clearly shows that the debris from an air burst is potentially fractionated, even though the debris taken as a whole is obviously representative. In order to actualize this potentiality, a particle separation process must occur. If the separation is the result of unequal rates of sedimentation through the atmosphere, the process could extend over long periods of time, and the degree of actualization would increase as time went on.

The correlation of radiochemical data from fractionated samples of air-burst debris throws light on the fundamental nature of the processes involved in debris formation: the nuclear processes of the primary fission products; the dispersion and subsequent nucleation, condensation and agglomeration of the carrier material; the incorporation of the primary fission products in the particle or their deposition on the particle surface. The correlative parameters show the influence of matrix material on the character of the fractionation and provide input data for prediction schemes, such as the radial-distribution model.[3]

A search of available radiochemical data on air-burst debris revealed thirteen detonations in which factors of ten or more were observed between the same radionuclide ratios in different samples from the same detonation. Two additional shots were included to extend the range

*Benson, P., Gleit, C. E. and Leventhal, L., "Physical Characteristics of Single Particles From High Yield Air Bursts," and "Radiochemical Fractionation Characteristics of Single Particles From High Yield Air Bursts," to appear in the Proceedings of the Second USAEC Symposium on Radioactive Fallout From Nuclear Weapons Tests, 1963.

of the weapon yields. These detonations ranged in total yield over a factor of about 300. All the radiochemical data available from these fifteen detonations were used in this work. For each of these detonations, the investigators in charge of the analyses had estimated representative (i.e., unfractionated) values of the radionuclide ratios obtained. Therefore, the ratios of fractionated radionuclides were converted to fractionation ratios ($r_{i,j}$) by simply dividing them by the representative values, as described in Reference 4.

Standard statistical methods were used to analyze the correlations between ratios, to test the variation of the correlation parameters with total weapon yield, and to test the hypotheses that nuclides of certain groups do not fractionate from one another.

RESULTS

To illustrate the range of fractionation observed in air-burst debris, Table 1 lists for each shot the ratio of the largest $r_{89,95}$ value observed to the smallest $r_{89,95}$ value observed. The shot numbers are assigned in order of increasing yield. The data shows that air-burst debris can be extremely fractionated. There is no evident trend of the ratio with yield. The wide variation in the ratio probably manifests nothing more than its dependence upon the sampling conditions. The observed $r_{89,95}$ values ranged from 0.002 to 5.3, although few fell outside the range of 0.1 to 3.0.

The next property considered was the relative ability of logarithmic and linear correlations to fit the data. The measure used for goodness-of-fit was the coefficient of determination (i.e., the square of the coefficient of correlation).[5] These coefficients indicate the fraction of variance in the dependent variable accounted for by the best fit of the type indicated. Individual shots varied considerably and were about equally divided in their preference. Table 2 shows the cumulative coefficients (i.e., obtained from treating all the data from all shots as belonging to a single population) for several important radionuclides which fractionate from Zr^{95} to varying degrees. With the single exception of Ce^{141}, which shows no significant difference, the cumulative coefficients indicate a preference for the logarithmic correlations. Unfortunately, this preference is influenced to some unknown extent by the type of scatter in the data. Data which have about the same relative uncertainty (as these do) will tend to follow a logarithmic correlation, while data which have about the same absolute uncertainty will tend to follow a linear correlation.[6]

TABLE 1

Range of Observed $r_{89,95}$ Values for Air Bursts

Shot Number	$(r_{89,95})$ max/$(r_{89,95})$ min
1	7
2	22
3	19
4	28
5	26
6	919
7	43
8	64
9	70
10	449
11	10
12	90
13	19
14	503
15	5

TABLE 2

Cumulative Coefficients of Determination for Important Fractionation-Sensitive Nuclides

Radionuclide	Coefficient of Determination	
	Logarithmic Fit	Linear Fit
Sr^{90}	0.71	0.51
Y^{91}	0.85	0.60
Cs^{137}	0.69	0.60
Ba^{140}	(0.76)*	(0.65)*
Ce^{141}	0.71	0.72
Nd^{147}	0.37	0.05
Np^{239}	0.78	0.64

*Slopes are yield dependent.

TABLE 3

Regression Slopes for Statistically Significant Cases of Yield Dependence

Radionuclide	Correlation with W (kt^{-1})	Correlation with Log W
Y^{91}	0.00016 ± 0.00015	-*
Mo^{99}	0.00019 ± 0.00011	0.066 ± 0.039
Ba^{140}	0.00016 ± 0.00009	0.053 ± 0.035
Ce^{141}	0.00011 ± 0.00007	-*
Nd^{147}	-0.00013 ± 0.00010	-*

*Slopes did not exceed confidence limits.

The yield dependence of the slopes of the logarithmic correlation lines were calculated for the nuclides Sr^{90}, Y^{91}, Zr^{97}, Mo^{99}, Cs^{137}, Ba^{140}, Ce^{141}, Nd^{147} and Np^{239}. This was done for both total device yield W and for log W. In most cases the regression slope for this dependence was equal to or less than the 95 % confidence limit. The results for those cases where the regression slopes exceeded the confidence limits are shown in Table 3. The Ba^{140} slopes appear to be definitely yield dependent, the dependence is in the expected direction, and the slopes themselves varied from 0.41 to 0.92. The Mo^{99} slopes also appear to be yield dependent. These ranged from 0.10 to 0.59. The cases for the other nuclides appear much less definite.

We come now to the values of the regression slopes for logarithmic correlations of the data from all shots treated as a single population. Because of our basis of presentation, these slopes are measures of the volatility of behavior of the various mass chains. The cumulative slopes for the logarithmic correlations, their 95 % confidence limits, and the number of data points involved, are shown for various important radionuclides in Table 4. Nuclides are listed in order of decreasing volatility (decreasing slope) for air-bursts. Although not strictly admissable because of yield dependence, Mo^{99} and Ba^{140} have been included to indicate typical values for these cases. Listed for comparison are the cumulative results from high-yield surface bursts which have been taken from Reference 4 and converted to the present basis (i.e., ratios are here based on Zr^{95} instead of Sr^{89}).

Table 5 shows the slopes of log $r_{i,j}$ vs log $r_{89,95}$ for various nuclide pairs with similar behavior.

TABLE 4

Cumulative Slopes for Various Important Radionuclides

Radionuclide	No. of Data Points	Cumulative Slope	High-Yield Surface Bursts
Sr^{89}	By definition	1.000	1.000
Sr^{90}	209	0.92 ± 0.08	0.69 ± 0.08
Cs^{137}	190	0.90 ± 0.09	1.07 ± 0.17
Np^{239}	280	0.80 ± 0.05	-0.05 ± 0.09
Cs^{136}	194	0.67 ± 0.08	-
Ba^{140}	301	(0.62 ± 0.04)	0.57 ± 0.09
Y^{91}	192	0.56 ± 0.03	-
Ce^{141}	232	0.40 ± 0.03	-
Mo^{99}	300	(0.30 ± 0.05)	-0.10 ± 0.09
Nd^{147}	193	0.22 ± 0.04	-
Zr^{95}	By definition	0.000	0.000

TABLE 5

Slopes of log $r_{i,j}$ vs log $r_{89,95}$ for Cases of Similar Behavior

Nuclide i	Nuclide j	No. of Points	Slope
Sr^{90}	Sr^{89}	209	-0.08 ± 0.08
Cs^{137}		190	-0.10 ± 0.10
Cs^{137}	Sr^{90}	150	0.00 ± 0.11
Ag^{112}		132	-0.05 ± 0.16
U^{240}	Np^{239}	185	0.00 ± 0.02
U^{237}		279	0.00 ± 0.17
Ag^{111}		250	0.00 ± 0.02
Cd^{115}		229	0.00 ± 0.04
Cd^{115m}		214	-0.09 ± 0.08
Cs^{136}	Ba^{140}	190	0.04 ± 0.08
Pu^{238}	Mo^{99}	85	-0.01 ± 0.12
Pu^{238}	Nd^{147}	70	-0.09 ± 0.09
Ce^{144}		164	-0.05 ± 0.20
Pr^{143}		188	-0.10 ± 0.03
Tb^{161}	Zr^{95}	156	0.04 ± 0.05
Sm^{153}		159	0.03 ± 0.05
Eu^{156}		175	0.00 ± 0.05
Zr^{97}		239	-0.03 ± 0.03

DISCUSSION

The simplified picture of fallout formation has visualized a yield-dependent time of condensation or solidification of debris, at which refractory elements were largely incorporated in the particle matrix while volatile elements had to either cool further or decay to refractory elements before they could condense on the available surfaces. Some of the results in the preceding section are unexpected from such a model, but it cannot be universally stated whether this disagreement lies with the fault of the model, of the basic nuclear data and its application, or in the accuracy and the treatment of the radiochemical data. Each anomaly must be treated individually, compared with other correlations, and the relative contribution of all four possible causes evaluated.

The insensitivity of correlation parameters to yield, in all cases except Mo^{99} and Ba^{140}, simplifies the prediction of debris properties but is difficult to reconcile with the simplified model described above. Equally difficult to explain on a physical basis is why these particular nuclides should show the greatest yield dependence. Plots of correlation parameter vs. yield show a great deal of scatter and the results here may simply reflect a larger number of data points (cf. Table 4) and more reliable results for these nuclides, rather than signifying a real insensitivity for the others.

The observed volatility of Np^{239}, on the other hand, appears more real. Besides the magnitude of the volatility, the results are well supported by independent data. Thus, Np^{239}, U^{237} and U^{240} are all present as uranium at condensation times and, as Table 5 shows, the data from these three nuclides are all in excellent agreement. Moreover, a similar, though less marked, behavior has been reported for the silicate surface bursts Small Boy and Johnie Boy.*

In using Table 5 to determine whether two nuclides do not fractionate from each other, the slope, confidence limits, and the chemical and nuclear properties of the precursors must all be considered. The following groups of nuclides fulfill all these qualifications well: U^{237}, Np^{239} and U^{240}; Ag^{111} and Cd^{115}; Zr^{95} and Zr^{97}; and Sm^{153}, Eu^{156}, and Tb^{161}. The data also indicate that there is no reason for excluding

*Crocker, G. R., Kawahara, F. K., and Freiling, E. C., "Radiochemical Data Correlations on Debris From Silicate Bursts," to appear in the Proceedings of the Second USAEC Symposium on Radioactive Fallout From Nuclear Weapons Tests, 1963.

Ag^{112} from the silver group, that the silver group does not fractionate sensibly from the uranium group and that the zirconium group does not fractionate from the heavy rare earth group. As would be expected, Cs^{136} and Ba^{140} behave very similarly. The volatile nuclides Sr^{89}, Sr^{90} and Cs^{137} may also follow one another in air-burst debris and Cd^{115m} may follow Cd^{115} but the indication that they do is marginal, to say the least. The data of Tables 4 and 5 indicate that Pr^{143} is more volatilely behaving than the heavy rare earths and that Nd^{147} behaves more volatilely than Pr^{143}. The confidence limits for Ce^{144} are too wide to establish its position among the other rare earths.

Figure 1 presents a visual comparison of the chain volatility inferred from fractionation behavior with the parent thermodynamic volatility shown by Fig. 2 of Reference 7. In the latter, Bedford and Jackson show the predominant form of the condensed phase and its equivalent pressure at 2500°K and 1 atm O_2 for the fission-product elements and uranium. The correspondence between the two sets of data indicates that the thermodynamic equilibrium model for nuclear debris formation proposed by Miller[8] may be applicable to air bursts and this possibility should be investigated further.

Some of the results here were compared with those for silicate surface bursts in the work of Crocker, et al., cited above. The comparison with results from high-yield surface bursts in Table 4 shows Np^{239} to be the nuclide whose behavior differs most in these environments. The difference between the values for the 140 chain in the two cases is well within the 95 % confidence limits as is the difference between values for the 137 chain. Discussion of the differences for Sr^{90} and Mo^{99} are best postponed until work in progress on additional correlations from high-yield surface bursts in the Pacific area is complete.

In conclusion, it is evident that our understanding of the situations prevailing in the formation of air-burst debris is rudimentary in all phases: the basic theory is lacking; the models are crude and it is somewhat surprising that they work as well as they do; fundamental thermodynamic and nuclear data are unavailable; the environmental conditions during debris formation are inadequately defined; the particles are formed and grow and coagulate at different times, places and temperatures, the larger ones being probably the first formed; and the subtlety of some of the effects sought appears to be beyond the accuracy of the data and the methods of correlation. For these reasons, no more detailed treatment has appeared to be warranted at this time. As correlation data accumulate from other burst types, as fundamental data becomes available, and as basic processes are better understood, more sophisticated statistical treatment and interpretation may prove fruitful.

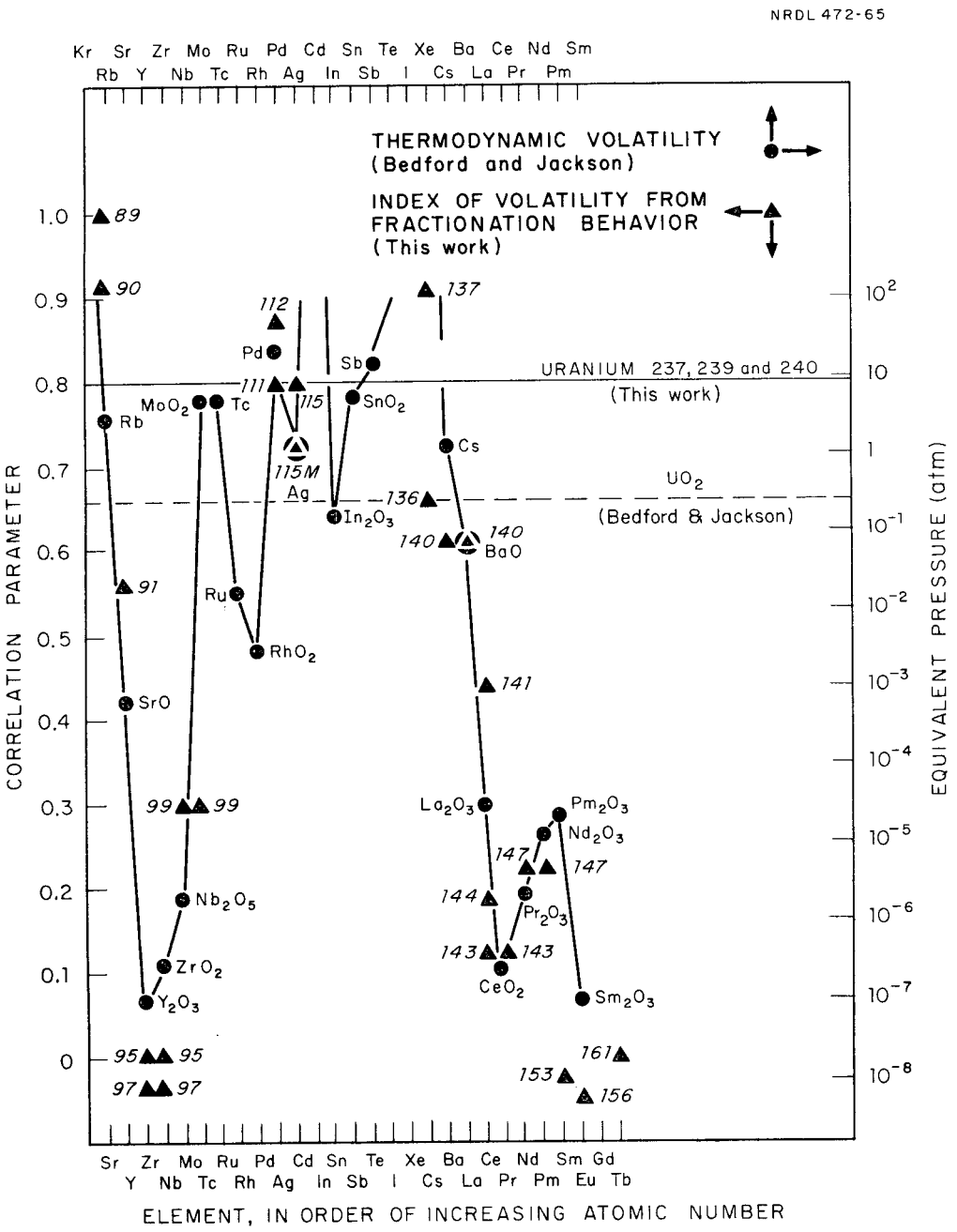

Fig. 1 A Comparison of Chain Volatility Inferred From Fractionation Behavior with the Thermodynamic Volatility of the Parent.

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